MODEL STUDIES ON NITROGEN AND SULPHUR TRANSPORT AND DEPOSITION SINCE 1993 IN THE BALTIC SEA SURROUNDINGS

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Abstract: Deposition and concentrations of nitrogen and sulphur compounds, calculated with the model Hilatar over the resent years, are presented. The model structure, performance and development of its parameterization schemes are shortly reviewed. Model applications in estimating background area pollutant concentrations and depositions since 2000 and with simultaneous development of emissions, as well as trends in the airborne NO_X load to the Baltic Sea and the effect of the specific AIS ship emission and North-West Russian emission inventories in comparison with use of EMEP emissions are discussed.

Key words: *airborne deposition, nitrogen, sulphur, regional modelling.*

1. TOOLS AND METHODS

Model

Hilatar (Hongisto 2003a,b, http://www.fmi.fi/research_air/air_25.html) is a nested Eulerian mesoscale air pollution transport model of the Finnish Meteorological Institute, in which the time change of concentrations are calculated by numerically solving the transport equation containing terms for emissions, advection, vertical turbulence, chemical transformation, dry deposition and scavenging by rain. The model calculates gaseous and particle concentrations and deposition (dry and wet) for NO(g), NO_x(g), HNO_3(g), NO_3(p), PAN(g), NH_4NO_3(p), NH_3(g), SO_2(g), SO_4(p) and $(NH_4)_{1.5}SO_4$(p). The model covers Europe or it's sub-areas with varying grid resolution (0.5^o–0.08^o). The high-resolution sub-model is linked to European model by adding the long-range transported compounds to the air flowing in at the sub-model boundaries. Hilatar has been validated by model-measurement inter-comparisons using monthly, weekly or daily measurement data (concentrations in air and precipitation) of over 90 European EMEP-stations (www.emep.int), Hongisto et al., 2003) using measurements from field campaigns (including ship and coastal measurements over the Baltic Sea, (Schulz et al., 1999) and by model-model inter-comparison (Zlatev et al., 2001).

Input data

The model is accompanied with a meteorological data base containing pre-processed time series of three-dimensional 6th hour HIRLAM forecasts over the Baltic Sea area since 1993, and over Europe since 1995. The European model uses gridded EMEP-webdab emissions which were corrected for selected North-Western Russian areas using information of the old and on-going common Finnish-Russian projects concerning environment in the Border area (Pellikka et al., 2006, Ekimov et al., 2001, Varkonyi et al., 2008, AMAP-reports, old FMI-SRI Atmosfera inventory and information on Statistical Centre of Karelian Republic). The high-resolution model uses Finnish stack- and area-source emissions, specific inventory for Kola, Karelia, Leningrad Oblast and St. Petersburg and EMEP emissions over other areas. Over the Baltic Sea Hilatar model uses ShipNODep (http://www.shipnodeff.org/ Jalkanen-Kalli in HARMO 12) international ship inventory, where NOx emissions of individual ships are estimated using information on ship direction, speed etc. from the decoded AIS, Automatic Information Signals, connected to Lloyds data base for ship machinery data. There was assumed a linear dependence between the SOx emission and NOx emission. The conversion factor from NOx emission to SOx emission is 0.40, corresponding to new S-content regulations for the fuel used in international traffic over the Baltic Sea.

Emission trends in the main research areas, Finland and countries surrounding the Baltic Sea, show no significant reductions in the oxidized and reduced nitrogen emissions since 2000, as seen from Fig. 1. NO_X emissions of the contries having largest contribution to the deposition to the Baltic Sea decreased from 21.2 Mt NO2 in 1990 to 16.9 Mt in 1995 and 14.8 Mt in 2000, staying at the same level until 2005 and varying mainly due to meteorological reasons. SO_2-emissions of the main countries contributing to the deposition to Finland reduced from 24.1 Mt SO_2 to 12.6 Mt SO_2 between 1990 and 2000, decreasing slightly still after that down to 11.3 Mt in 2005. NH_3 emissions of the same countries did decrease to 76 % from 1990 to 2000, and stayed at the 2000 level afterwards.

Russian Emission inventory

Although after the collapse of the Soviet economy system since beginning on 1990's dramatic changes in industrial production volumes and, consequently, reductions in emissions throughout Russia occurred, the emission reductions are not as big than that in the EMEP data base www.emep.int (Fig 2.) According to Russian sources sulphur dioxide and heavy metals emissions from the Kola Peninsula cupro-nickel industries fell from 586.3 t to 257.5 t between 1985 and 2003. The emissions of Pechenganikel decreased from a maximum of over 400 kt SO2 in 1977 to 110.5 kt in 2004 (AMAP) and 106.8 kt in 2005 (Karri Eloheimo, 2007). The emissions of the Republik of Karelia have decreased during the last 20 years especially due to the natural gas pipe line, there are about 9 000 point sources, and gas and particle cleaning units are available in 2 000 sources and about 52,2 % of all emissions are treated. Biggest enterprises emitting polluting substances are Kostamus, Kontupohja, Segesha (pulp and paper mill), aluminium smelter and Pitkäranta (cellulose factory). AIS emissions compared to EMEP Baltic Sea emission inventory are presented in Figure 3.

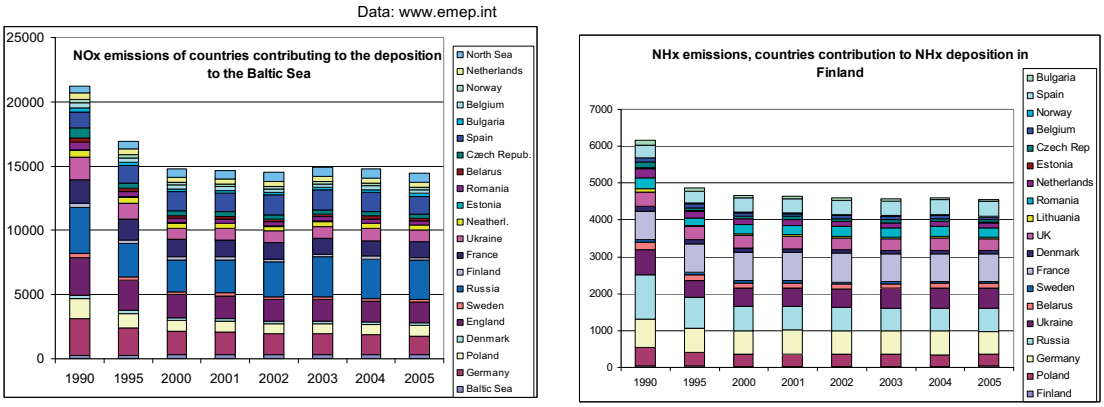

Figure 1. Nitrogen emission trends in the EMEP inventory of countries contributing to deposition to the Baltic sea / Finland.

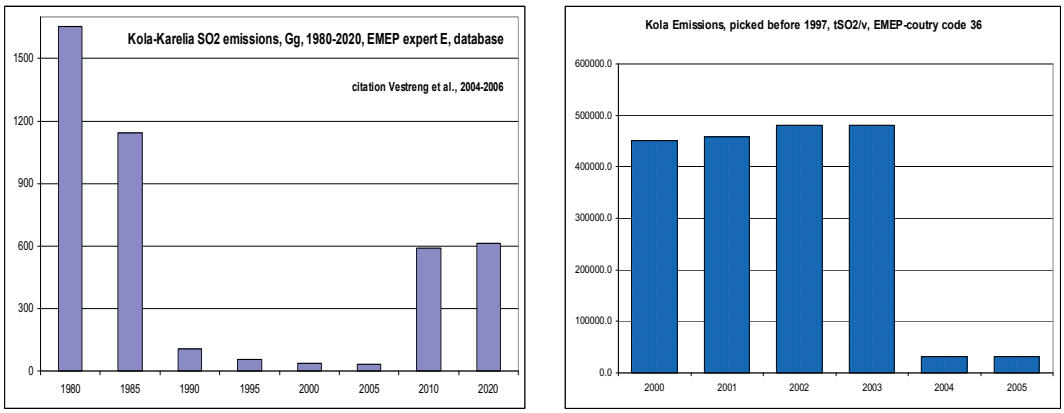

Figure 2. left: Current state of the Russian emissions in ww.emep.int (Spring 2008); right: Situation in 2004 and 2005 compared to annual inventories before 2003; in 2004 all sulphur emissions in Kola-Karelia were reduced by a factor seen in the Figure.

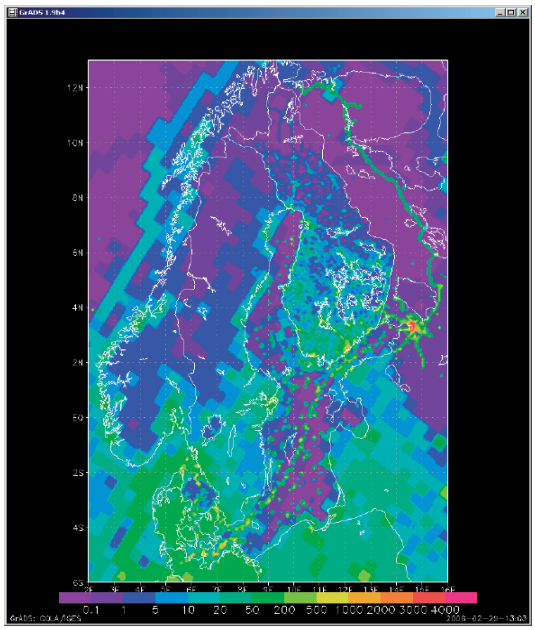

Figure 3. Instant ship emission intensity compared to land-based emissions, March 2006, NO2, molecules $m^{-3}s^{-1}\bullet e^{-11}$.

2. RESULTS

Simulated SO_2, NO_X and NH_3 depositions in 2005 and 2006 in Europe are presented in Figure 4, NO_X deposition caused by ship emissions ($mgNm^{-2}$), calculated as the difference between the modelled deposition with and without the NO_X emissions from shipping in the period 1.3.2006-28.2.2007 in Fig 5 and sulphur concentrations and deposition caused by the Baltic ship traffic in Figure 6. The latest EMEP-emission inventory was for the year 2005. Annual depositions to the Baltic Sea are higher however over other areas they do not differ much in comparison with the EMEP-model calculated depositions corresponding to the similar structure of the models, except near the Finnish-Russian border areas. Slight differences due to annual meteorological variation can be seen in the 2005-2006 depositions. The annual NO_X deposition to the Baltic Sea decreased from around 178 kt to 140 kt from 1993 to 1997, but increased to 180 kt in 1998 (Hongisto and Joffre 2005). In 2000 we get 191 kt with a model with increased vertical and horizontal resolution and slightly updated chemistry, and the NO_X deposition varied from 141 kt (2005) to 190 kt (2001) since then. There occurred no significant emission reductions, variation is caused mainly due to meteorological differences between the years. In comparison to EMEP deposition to the Baltic Sea, 114.3 kt N 2005, Hilatar deposition is higher, but according to Tarrason et al., 2007, NO_X wet deposition is underestimated in the EMEP calculations on average by 32% in Europe, however the underestimation is bigger at some stations at the coast to the Baltic Sea. The total nitrogen deposition to the Baltic Sea decreased by 22% between 2000 and 2005 according to Hilatar and by 23% according to EMEP calculations, however, end of year 2000 was very rainy over the Baltic Sea, and the October-December deposition in 2000 was exceptional high.

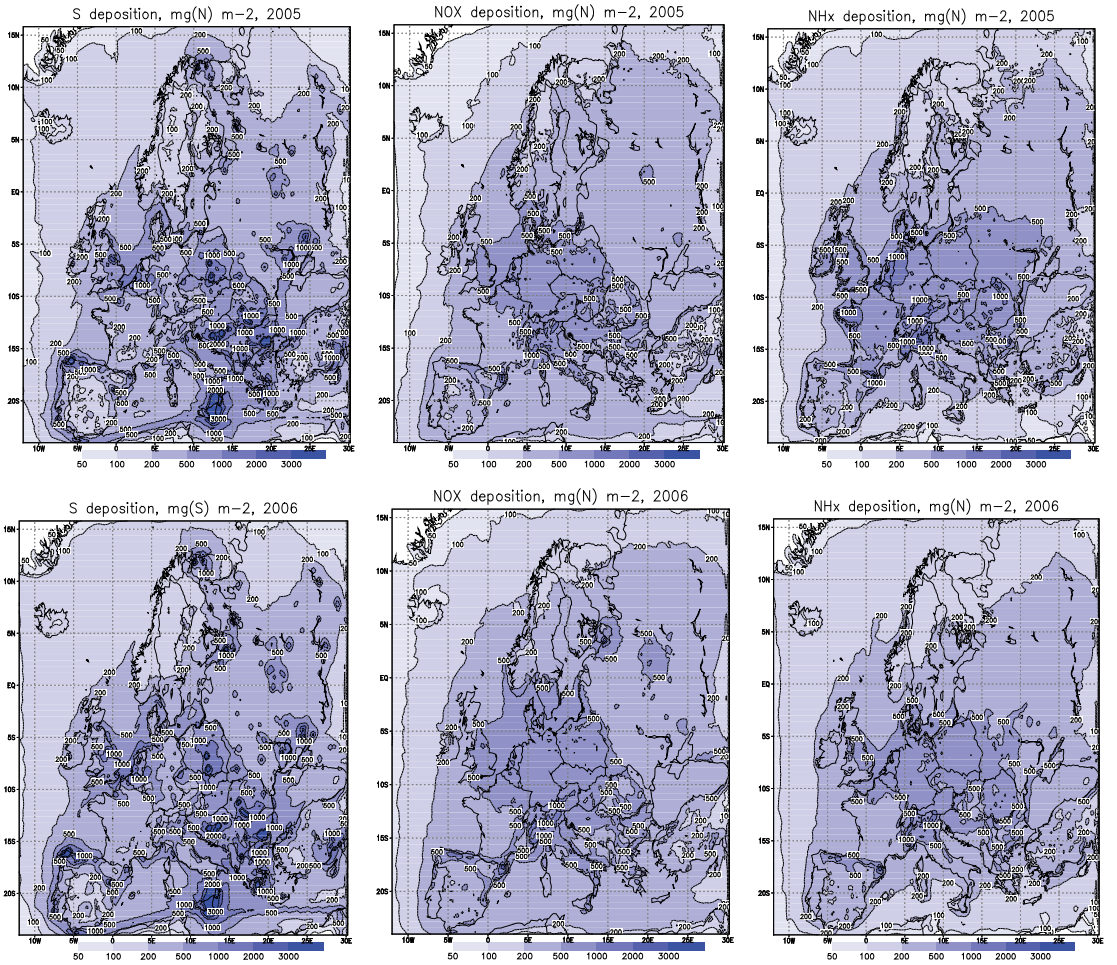

Figure 4. European deposition sums in 2005 and 2006, mg(N) m^{-2} or mg(S) $m^{-2.}$

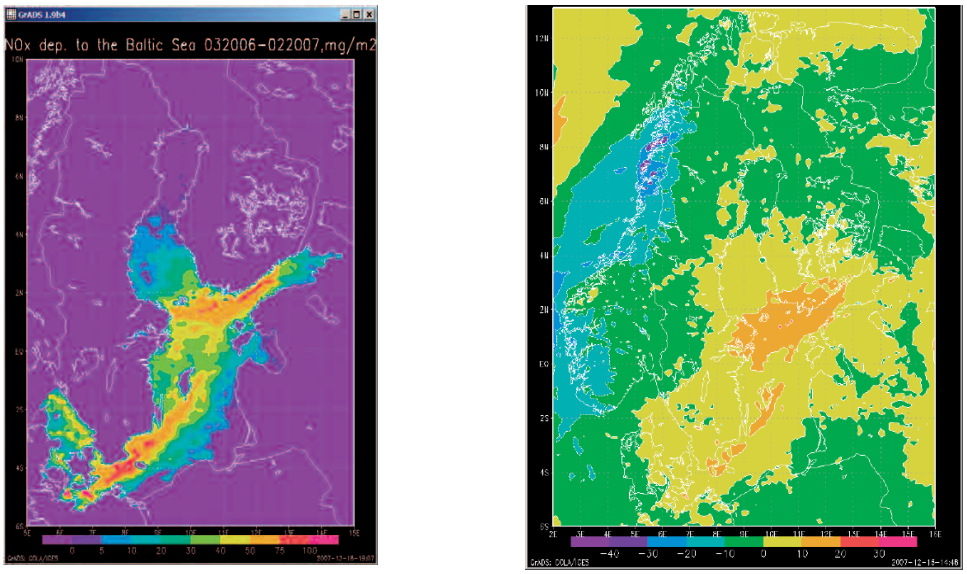

Figure 5. Left: Annual NOx deposition caused by ship emissions (mgN/m^2). Calculated as the difference between the modelled deposition with and without the NO_X emissions from shipping in the period 1.3.2006-28.2.2007. Right: Percentage of annual NO_X deposition caused by ship emissions

Due to seasonal variation of chemical transformation speed, life time and transport distance of NOx compounds in atmosphere as well as seasonal variation of land-base emissions, precipitation, dry deposition velocities, average wind velocity and turbulence and stability of the atmospheric boundary layer, the relative effect of the shipping emissions on the deposition vary widely with season. In the mid-summer (when the marine ecosystem in the Baltic Sea is most receptive to external nutrient input), locally up to 50% of the monthly NOx-deposition in the northern Baltic proper can originate from shipping.

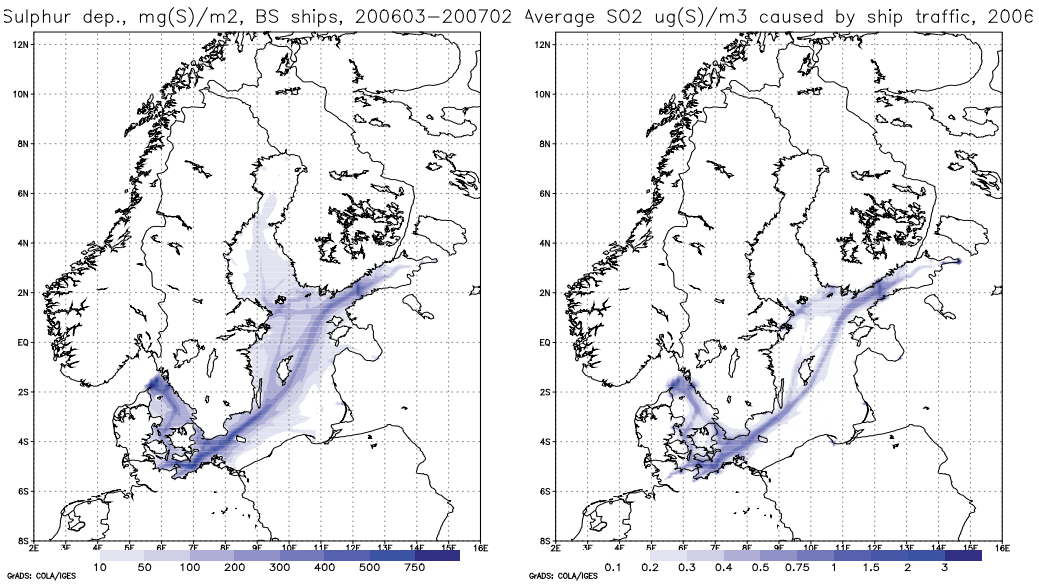

Figure 6. Sulphur deposition and average SO2 concentrations caused by the Baltic ship traffic.

3. CONCLUSIONS

Baltic ship emissions have been increasing and their importance for the eutrophication of the Baltic Sea in summer might be more important than previously expected. The impact to the Baltic Sea deposition of the emissions development and meteorology since 1993 will further be studied. The contribution of the Nort-west Russian emissions to the air quality in Lapland is still remarkable, although the problem will disappear partly due to modernization of the Petshenganikel metal industry by Norwegian-Russian co-operation, which is currently carried out.

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